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PHONON REFLECTION AT NOBLE GAS INTERFACES.(U)

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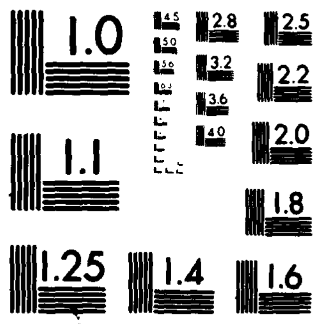

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(6) Phonon Reflection at Noble Gas Interfaces,

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ABSTRACT: High resolution phonon reflection spectra from thin films of noble gases condensed onto a sapphire substrate at 2 K are presented. Phonon reflection signals from films of helium, neon, argon, krypton and xenon are found to be essentially identical, suggesting that quantum effects play no role. We argue, however, that the conventional interpretation of a change in the signal as due to heat escaping from the system cannot be valid for the solidified rare gases.

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Heat pulse experiments which use thin film ohmic heaters as generators and superconducting bolometers as detectors of thermal frequency phonons have become an important method of investigating phonon reflection from interfaces. One of the principle motivations for studying this problem is to gain some understanding of the microscopic mechanism of the well-known anomalous Kapitza conductance at solid/liquid helium interfaces. The Kapitza effect may be studied in a phonon reflection experiment by comparing the bolometer signal due to reflection from a vacuum interface with the signal when helium is present.(1) The difference between the two signals is attributed to energy transmitted through the interface. The decrease in the reflection signal which is usually(2) observed when helium is introduced is hundreds of times larger than expected from elastic theory. In contrast, if neon rather than helium is placed at the interface, the decrease in the signal is roughly as expected from elastic theory.(3) Because of this observation and the arguments presented with it, it is now widely believed that the anomalous Kapitza effect occurs only in quantum media.

In this paper we shall first show that, to the extent that the signal from a helium interface is anomalous, precisely the same anomaly is found for all the noble gases. Second, we show that the missing signal cannot simply be attributed to energy that has been transmitted through the interface and carried away. The differences between our conclusions and those of previous investigators are a consequence of the very high time resolution in the data we have obtained.

The apparatus used in these investigations is similar to one described in detail previously.(4) The phonons are propagated through a cylindrical sapphire crystal (10 mm x 57 mm dia) which is oriented so that the three-fold

C axis and the two-fold X axis are parallel to the cylindrical faces. The crystal surfaces are mechanically polished. Thermal phonons are generated by an aluminum heater film (100 nm thick) and detected with a superconducting tin bolometer film (200 nm thick) which is biased in the linear portion of its transition in a magnetic field. The crystal is immersed in a superfluid bath at 2 K. The reflecting surface is initially in vacuum but can be covered by a film of liquid helium, or a variety of solidified gases. The gas inlet tube is thermally insulated and heated to prevent the gases from freezing out before condensing on the sapphire surface.

If the heater and bolometer are displaced along the X axis, the reflection signal is as shown in Fig. 1. Several sharp peaks as well as a broad superimposed diffuse peak can be clearly seen. The sharp peaks in the reflection signal are due to specular ($k_{||}$ conserving) processes which have a well-defined arrival time that can be accurately predicted from anisotropic elastic theory,[4] while the diffuse signal, which is probably due to surface roughness, has a broad distribution of arrival times. The magnitude of the diffuse signal is very sensitive to the relative orientation of generator and detector with respect to the crystal axes. As described in refs. (5 & 6), the configuration of Fig. 1 leads to a large bulk scattering background and a particularly large diffuse reflection signal because the caustic surfaces associated with the heater and bolometer intersect.

The effect of admitting helium to the reflection surface is shown in the dashed curve in the figure. The most striking effect is the decrease of the diffuse peak. The diffuse scattering signal is observed to be drastically reduced in all geometries. In fact, a careful analysis[6] suggests that the entire effect of helium is to remove the diffuse signal, while the specular signal is changed only little, if at all.

Figure 2 shows a comparison of the vacuum reflection signal and the argon interface reflection signal for precisely the same geometry as in Fig. 1. The signals shown in Figs. 1 and 2 are in fact identical to within the 3% reproducibility of the experiment. The exact thickness of the solid argon film deposited on the sapphire is unknown, but is certainly less than 1 μm. The possibility of helium leaks or contamination was tested by pumping on the cell with a helium leak detector after the argon had been deposited.

This experiment was repeated with films of neon, krypton and xenon, with a number of different heater-bolometer geometries. In each case the reflection signal from the sapphire/rare gas solid interface was essentially indistinguishable from a similar experiment with helium. This sequence of interfaces represents a variation of the speed of sound by a factor of 7 and a variation in density by a factor of 25.[(7)] Although it seems remarkable that the diffuse scattering signal is so insensitive to the acoustic parameters of the interface, standard elastic theory calculations of the phonon reflection coefficient assume reflection from a perfect planar interface and therefore should not really be expected to predict the behavior of the diffusely scattered phonons. The fact that the quantum zero point contribution to the energy in, e.g., argon or xenon, is completely negligible seems to indicate that the observed decrease in the diffuse reflection signal even in helium cannot be attributed to quantum effects.

It should be noted that these results are not inconsistent with previous phonon reflection experiments from helium and other classical solids. In ref. (3), the emphasis is on comparison of the observed reflection coefficients with predictions of elastic theory rather than with each other. Because the diffuse and specular portions of the signal were not resolved, it is less obvious that the same phenomena are occurring in, say, Ne as in He. Examination of the data in ref. (3) shows that the observed changes in the reflection signal for a sequence of quantum and classical interfaces differ by only a few percent. The approximate agreement between the data and acoustic mismatch theory for solid neon is fortuitous. If acoustic mismatch accurately described phonon reflection experiments from classical interfaces, one would expect more than a factor of 2 higher transmission into xenon than neon, which we do not observe. Rather than making comparisons to acoustic mismatch theory, we feel that the results of our experiments and the experiments reported in ref. (3) can best be understood by observing that the phonon reflection signal decreases by essentially the same amount irrespective of whether liquid helium, solid helium, D_2, H_2, Ne, Ar, Kr or Xe are deposited on the reflection surface.

For the case of helium films, there is a well-accepted explanation[8,9] for the decrease of the reflection signal. Some fraction of the phonons incident on the interface are absorbed into the film. The helium film cools by evaporation and is replenished by superflow or condensation from the vapor, so the total phonon energy reflected back into the crystal decreases. In this view, phonon reflection experiments with superconducting bolometers essentially measure the thermal conductivity of the helium vapor in equilibrium with the film. If there is insufficient vapor to conduct the heat away, the heat returns to the bolometer at a later time.[9]

Despite the plausibility of this model for the case of helium, it cannot account for results obtained for films of the rare gas solids. The latent heat of sublimation is 40 to 300 times higher in these solids than in helium, so evaporation is impossible with energies of 30 nj per pulse used in this experiment. Also, the vapor pressure of these solids at 2 K can be estimated to be less than 10^{-17} torr,[7] much too low to account for the observed decrease in the reflection signal. If a pulse of phonons is absorbed in a film of e.g., argon, it can relax to equilibrium only by re-emitting phonons back into the crystal. If the thermal energy is stored in the film for a time much longer than the phonon transit time through the film, which is approximately 1 nsec, we should observe its return at a later time. We have tested this possibility by monitoring the reflection signal for up to 100 μsec. If the bolometer signal is proportional to energy flux and the argon film sends all the energy back into the crystal eventually, conservation of energy demands that the time integral of the vacuum interface reflection signal and the argon film reflection signal should be equal. Figure 3 shows that even on long-time scales, the reflection signal from the argon film is always smaller than the vacuum signal. That result is a feature of all of our measurements, independent of the geometry of heater and bolometer. Thus the decrease in the reflection signal cannot be explained by a change in the angular distribution of the diffusely scattered phonons. Previous authors[9] have reported cases where energy does return at a later time from an adsorbed film. We have been unable to reproduce their results in spite of repeated attempts.

The only apparent way to reconcile our experimental results with the conservation of energy is to assume that the energy returns to the crystal in a form that is not detected by the bolometer. A likely possibility[9] is that the phonon pulse, which has a characteristic temperature of approximately 10 K, is thermalized by the argon film and the bolometer is not as sensitive to the thermalized heat pulse. One can test the frequency sensitivity of the bolometer over a narrow range by changing the heater power. A variation of heater power over two orders of magnitude changes the effective heater temperature by only a factor of 3, from, roughly, 5 K to 15 K.[10,11] Over this range we find the entire signal linear in the heater power, so the bolometer would have to become insensitive at frequencies below ≈ 200 Ghz to account for our observations.

Our results have important implications for the proper interpretation of phonon reflection experiments. Kapitza resistance studies using heat pulses are based on the fundamental assumption that the observed reflection coefficients from an interface are simply related to transmission coefficients by the conservation of energy. Our experiments show that this relationship is completely unreliable. The fact that the phonon reflection signal from a sapphire/helium interface is indistinguishable from the reflection signal from a sapphire/argon interface strongly suggests that most or all of the decrease observed for helium may not be due to heat transport through the helium vapor, but rather to whatever processes cause the decrease in argon and the other rare gas solids. One might even speculate that these interfaces have the same phonon reflection signal despite the large variation in acoustic properties because they are all anharmonic systems that can be expected to thermalize phonons effectively. Without a quantitative model to explain the decrease of the diffuse component of the reflection signal, the relationship of any reflection measurement to the steady-state Kapitza resistance is unclear.

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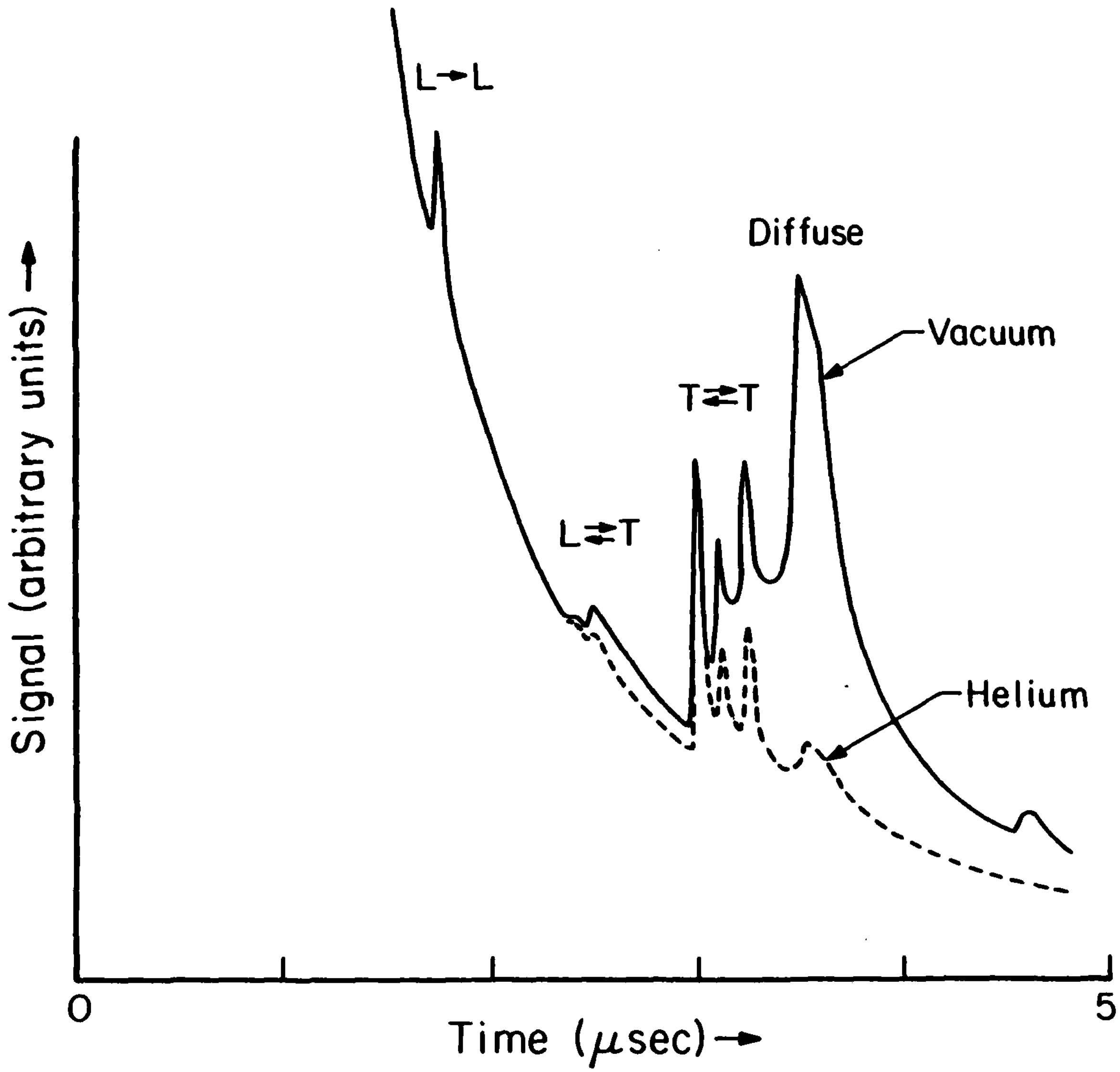
Figure Captions

Fig. 1. Bolometer signal as a function of time, showing specular peaks due to longitudinal (L) and transverse (T) phonons, and large broad peak due to diffuse scattering. Heater and bolometer are displaced along the X axis. The dashed curve shows the effect of introducing helium.

Fig. 2. Bolometer signal as a function of time for the same geometry as in Fig. 1. The dashed curve shows the effect of condensing a film of solid argon on the reflection surface.

Fig. 3. Long time behavior of reflection signal from vacuum and argon coated interface. Heater and bolometer are displaced along the C axis.



L→L
L⇄T
T⇄T
Diffuse
Vacuum
Helium
Signal (arbitrary units) →
0
5
Time (μsec) →

Figure 1

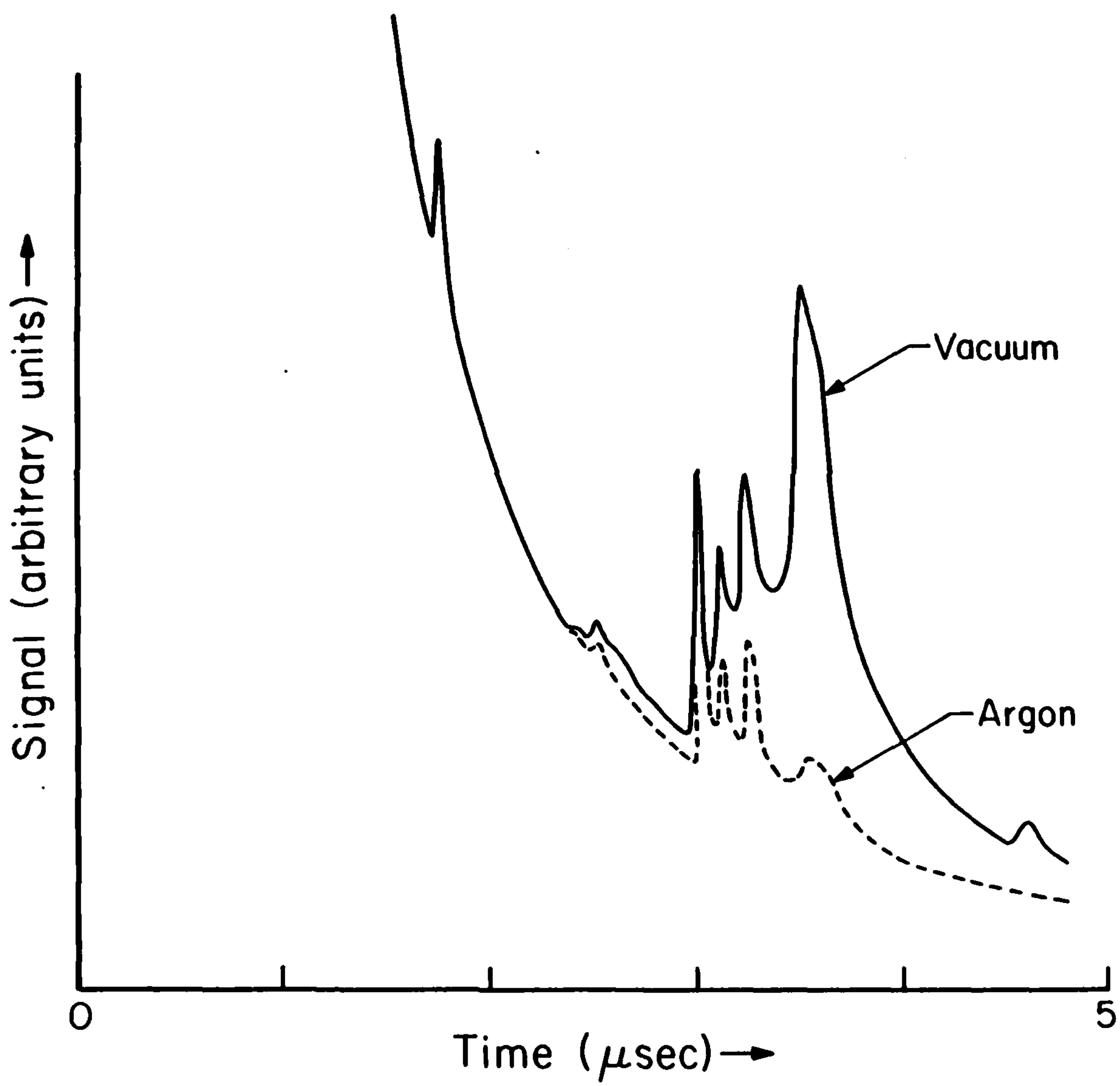


Figure 2

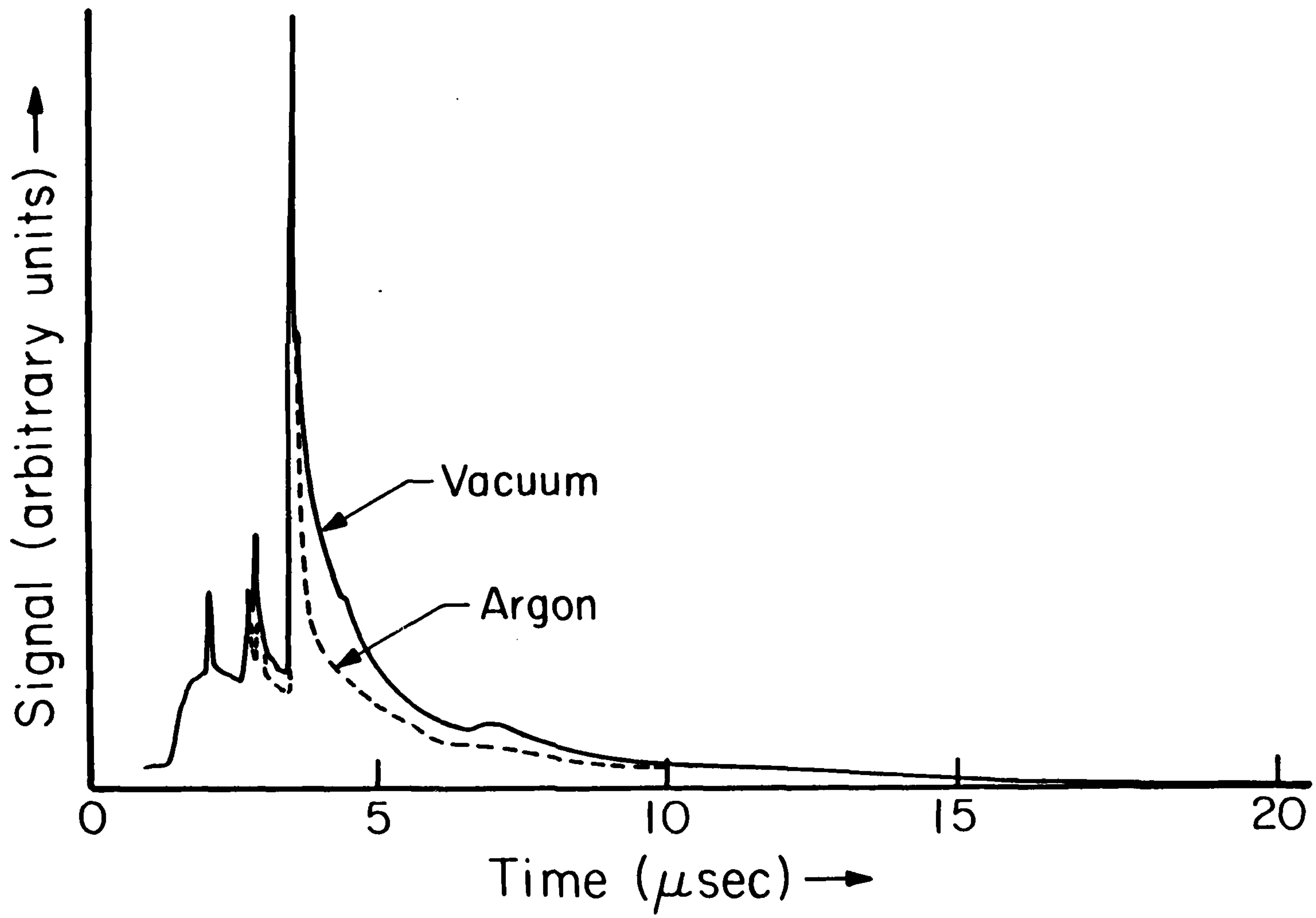


Figure 3

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